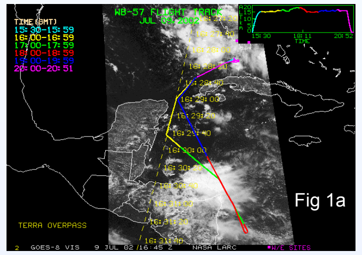

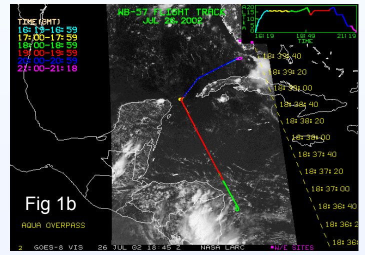

Observations of NO and NOy in the southernmost CRYSTAL-FACE mission flights

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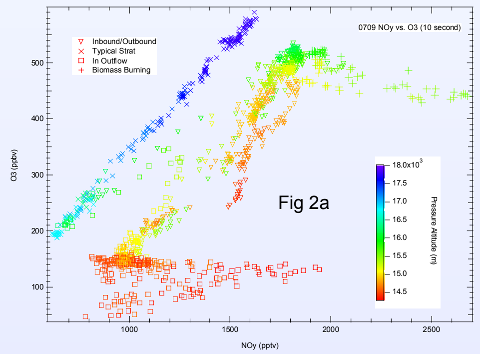

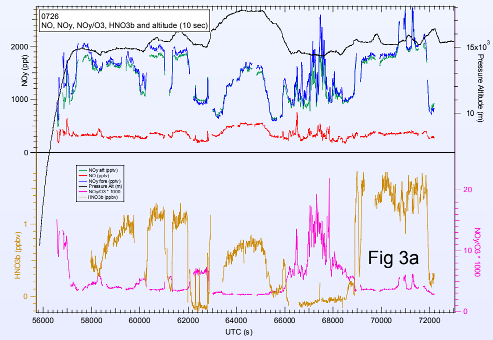

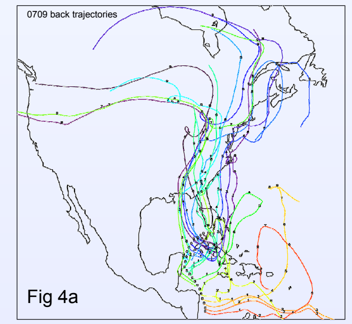

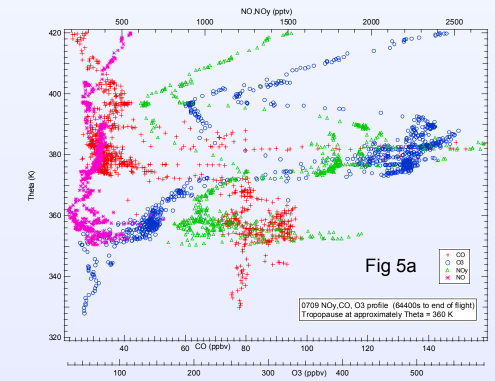

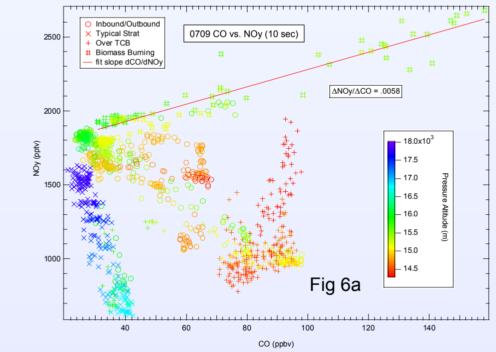

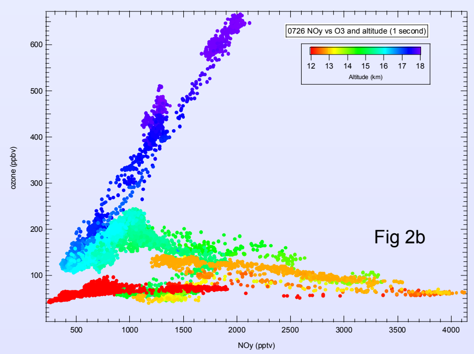

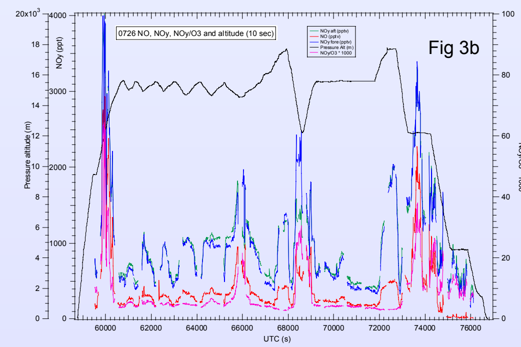

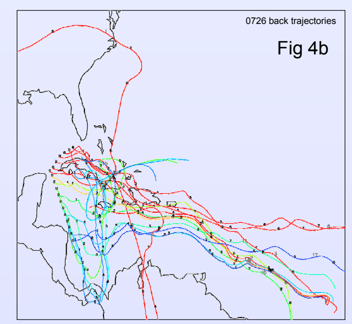

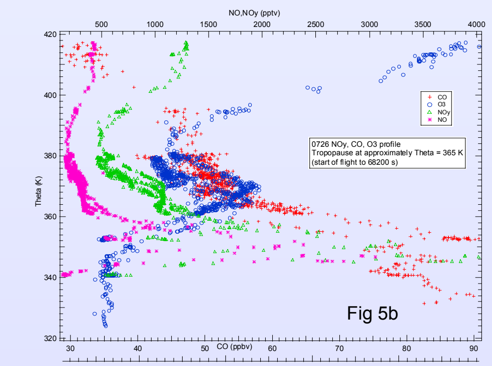

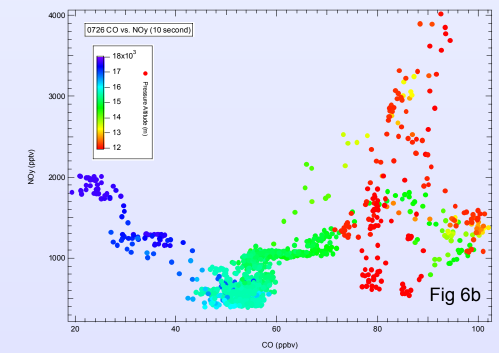

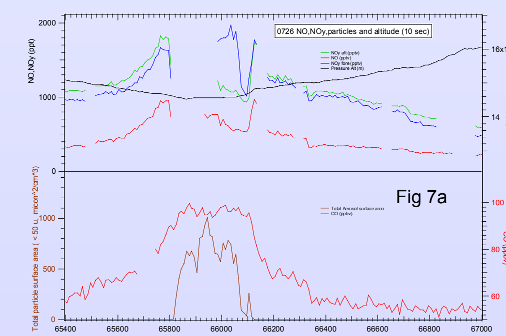

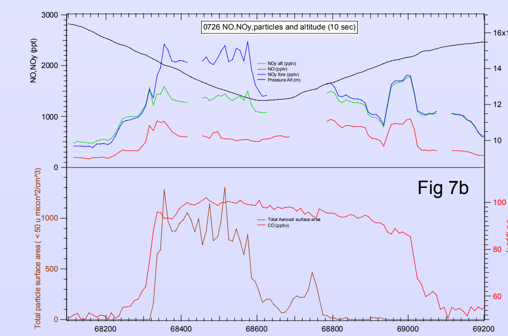

Introduction

Two southern flights of the WB-57 during the CRYSTAL-FACE campaign in Florida in July 2002 (Figs 1a & 1b) revealed contrasting sources of NO and NO_y in the UT and LS. While the flight tracks and altitudes were very similar between the two flights, air masses sampled traveled primarily from the North American Continent on 0709 and from low-latitude Atlantic sources on 0726. Convective influence from recent storm cells, clusters and residual outflow regions were measured on both flights. Outstanding differences in features between the two flights include unusual values of NO_y, Ozone and CO in the LS on 0709 compared to more typical LS constituent mixing ratios on 0726.

Flight Comparisons

0709 Flight

We divide the 0709 flight into four distinct segments based on the O_3/ NO_y correlation shown in Fig. 2a. These are labeled "Inbound/Outbound", "Typical Strat", "In Outflow" and "Biomass Burning". The "Typical Strat" and "In Outflow" events occurred over, or within, the anvil outflow of a large storm over the coast of Guatemala (Fig. 1a).

The "Typical Strat" segment was sampled when the aircraft flew over the system to about 17 km, providing the strong positive O_3/ NO_y correlation normally associated with lower latitude, lower stratospheric air. The aircraft then turned and descended to ~14.6 km, within the outflow of the storms. Increases in NO, NO_y, NO/ NO_y and NO_y /O_3 ratios (Fig. 3a) and particle surface area (not shown) confirm fresher convected air, possibly accompanied by lightning. Gas phase HNO_3 was near zero within this cirrus event.

The remainder of the flight was enroute to or from this convective system, in clear air, termed "Outbound/Inbound" in Fig 2a. A remarkable feature, termed "Biomass Burning", was encountered near 70600-71400 sec (Fig. 3a) while within the lower stratosphere. Unusually large CO (~160 ppbv) and larger NO_y and NO_y /O_3 ratios were observed in a layer approximately 0.5 km thick. The high CO has been attributed to injection of boreal forest fire emissions originating in Canada (see poster/talk by Jost et al.). Deep vertical mixing must have occurred as a result of frontal system activity or convection. Within this layer, no significant change in NO or HNO_3 occurred which suggests that the biomass plume was well aged chemically prior to vertical transport, and/or that HNO_3 was removed in the process. $\Delta NO_y/\Delta CO$ within the plume was ~0.006 (Fig. 6a), nearly identical to the average measured in aged boreal forest fire plumes by Wofsy et al. [1992].

Fig. 5a shows that the high CO mixing ratio layer was embedded within a much thicker layer of high O_3 mixing ratios (~550 ppb) between 340 and 400 K. Such large O_3 concentrations for this potential temperature range implies that high latitude, lower stratospheric air was advected south to the flight region. The back trajectories shown in Fig. 4a (only outbound legs plotted for clarity) corroborates this feature's source, and the possibility of aged biomass burning emissions impacting the flight region. We have not identified the event responsible for the injection into the lower stratosphere.

0726 flight

The correlation between O_3 and NO_y on 0726 (Fig 2b) is considerably simpler than the previous flight. There are basically two lobes, one very similar to that of 0709, and typical of the lower latitude, lower stratosphere. There is a second "tropospheric" lobe of large NO_y at relatively low O_3, measured from the samples taken on ascent from and descent into Key West.

Strong convective activity over Northern Cuba earlier in the day on 0726 brought LT air to 200 K cloud top temperature, at about 12 km, based on a Key West sounding earlier that day. Southeasterly flow at low altitude during the day indicated by wind profiles at Key West, and by visible GOES-8 images, suggests that sources of CO were not from anthropogenic sources in Florida but from Cuba. NO mixing ratios of 2-3 ppbv and NO_y mixing ratios of up to 4 ppbv are observed at approximately 12 km on climb-out and descent on 0726, well correlated to CO mixing ratios of 80-100ppbv. The large NO mixing ratios are very likely due to lightning activity in the storm (Fig 5b).

Ascent through 12 km occurred in approximately the same spatial location as the return descent through 12 km. Ascent values of 1-50 µm particles and water were considerably higher than the return sample values almost 4 hours later, with CO mixing ratios remaining similar. The NO/NO_y ratio also decreased from about .75 to about .6. GOES-8 IR images show small apparent motion of high altitude water features over the course of the flight, so that if we assume that ascent and descent sampled the same source airmass, these changes may be a result of chemical aging and ice particle evaporation. Profiles of CO, O_3, NO and NO_y on 0726 show increased mixing ratios from convection below the tropopause.

Two interesting cirrus encounters occurred on the 0726 flight, centered at approximately 66000 and 68500 seconds (Figs 7a & 7b). NO/ NO_y, CO, particles and water values, as well as GOES flight track images, indicate the aircraft was within anvil outflow. Larger NO and NO/ NO_y ratios of 0.6-0.7 suggest a contribution from lightning. (see poster by Weinheimer et al.)

Conclusion

The two southern flights were similar in flight path, altitude and time of day, but sampled dramatically different sources; Almost entirely low-latitude Atlantic on 0726 and almost entirely air over eastern North America on 0709. The most remarkable feature was the thick layer of larger mixing ratios of NO_y, O_3 and HNO_3 in the LS at low latitudes, which was advected from higher latitudes. Embedded within this feature was a layer of aged biomass pollution from the lower troposphere. Both flights sampled convective outflow from storms over Guatemala. On the 0726 flight, large NO mixing ratios, attributed to lightning produced in a storm over Cuba, were observed on ascent from and descent into Key West.

References

Wofsy, S.C., et al., Atmospheric Chemistry in the Arctic and Subarctic: Influence of Natural Fires, Industrial Emissions, and Stratospheric Inputs., *J. Geophys. Res.*, 97,16,731-16,746, 1992.